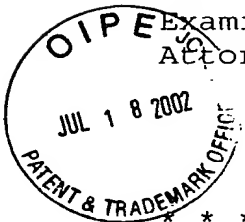


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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application : Bernard Reesink et al
Application No. : 09/744,134
Filed : 3/21/01
For : HYDROGENATION PROCESS
Examiner : Preisch, N.
Attorney's Docket : VER-140XX



Group Art Unit: 1764

I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Box Non-Fee, Assistant Commissioner for Patents, Washington, D.C. 20231 on 7-13-2.

By: 
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DECLARATION UNDER 37 CFR 1.132

Commissioner for Patents
Washington, D.C. 20231

Sir:

I, B.H. Reesink, citizen of The Netherlands, residing at Doorn, hereby declare as follows:

1. I am a co-inventor of the invention described and claimed in the above mentioned patent application.

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2. I have received training in Chemistry, having received the masters degree in Chemistry from the University of Utrecht.

3. I have had work experience in heterogeneous catalysis at Engelhard from 1986 through the present. I have been the author or co-author of approximately 3 technical articles, 7 patents and delivered papers relating to catalyst preparation and performance testing. I am a Senior Chemist in the Catalytic Materials department since 1986.

4. I understand that the Examiner for U.S. Patent Application Serial No. 09/744,134 has rejected claims of this application in part because he believes that the instant invention is obvious over Antos (US Patent No. 4,036,743) combined with Barre (US Patent No. 5,868,921).

5. Experiments were conducted and demonstrate that the results using the claimed process yield unexpected benefits. (See Attachment)

a. Evaluation of process of claim 1

The Attachment shows the preparation of catalysts A, B, C and D.

In the first experiment a catalyst A was compared with a catalyst system as is claimed in a process

according to claim 1 of the present invention. Catalyst system B (according to claim 1 of the invention) consisted three beds (one for each component). Since at the time of performing the experiment no SnO was available, FeMo oxide was used instead of SnO (in system B). It is however submitted that this is not regarded to be critical for the outcome of the results. The differences in concentration of the metals compared to catalyst A were compensated by diluting the FeMo bed and the Ni catalyst bed, such that a similar metal loading was obtained. Thus a proper comparison could be made.

Figure 1 clearly shows that system B has a superior activity performance. It can withstand a much higher sulfur dosage before the conversion is reduced. For example at a sulfur dosage of about 2.5 pulses/ml, a conversion of more than 90 % is achieved for system B under conditions wherein catalyst A only achieves about 20 % conversion.

From Figure 2, it can be concluded that the k-value in the catalytic reaction in a process according to the invention is not only higher, but maintains at a higher level after being exposed to sulphur.

Figure 3 shows the results of the effect of subsequent use in a hydrogenation of a sulfur contaminated feed stock of different catalyst systems, each containing the same amount of the metals. Catalyst A (similar to US patent 3,796,654), was the composite referred to above, comprising Pt, Sn and Ni. System C (according to claim 1, now with Sn, just like Catalyst A) and system D (according to claim 1, with Mo/Fe instead of Sn) both show a much higher conversion rate under the give reaction conditions.

Furthermore after 14 pulses, system D and particularly system C (containing Sn, like catalyst A) still show a significant activity, whereas comparative system A, has lost all activity after only 10 pulses.

Summarizing, these examples clearly illustrate that a system according to claim 1 of the present invention shows a surprisingly higher activity in hydrogenating an unsaturated compound than a catalyst according to the prior art. Furthermore, catalysts used in a process according to claim 1 show a much higher activity performance. As a result claim 1 is inventive.

B. Evaluation of process of claim 2

The results of a process according to claim 2 of the present invention are also shown in Figure 3. Catalyst system E shows a mixture of catalysts wherein the resulting amounts of Pt, Sn and Ni are the same as in the composite catalyst A according to US patent 3,796,654. Not only shows catalyst system E a much higher initial activity, but its activity does not decline during 9 consecutive feed pulses, whereas the activity of catalyst A gradually declines starting from the first pulse.

6. I declare further that all statements made herein to my knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardise the validity of the application or any patent issuing thereon.

18 June 2002

Date

271934

B.H. Roesink

[NAME OF PERSON SIGNING]

B.H. Roesink.

1,2,3,4, tetrahydronaphthalene (tetralin) conversion

Catalysts:

A catalyst according to US patent 3,796,654 was prepared. The catalyst was prepared using alumina as support material. The catalyst contained 0.375 wt.% Pt, 0.65 wt.% Sn and 5.0 wt.% Ni; PABD=0.33 g/ml.

Catalyst system B consists of 3 catalysts divided in 3 beds (in accordance with claim 1 of the present application). The first bed comprised a catalyst with 0.5 % Pt; PABD = 0.85 g/ml; the second bed comprised 4.1% FeMo; PABD = 0.5 g/ml; the third bed comprised a catalyst with 56 % Ni; PABD = 0.85 g/ml.

Hydrogenation performance:

The performance tests were carried out in a fixed bed with a tube diameter of 0,5" (HP 3) on equal catalyst volume basis using sieve fractions in the range of 0.15-0.25 mm. The catalyst volume of both systems was 5 ml. In order to obtain similar metal loading, the FeMo and the Ni catalyst of system B were diluted with inert particles.

After loading the catalyst system, the catalyst was activated by heating the reactor to 200°C under hydrogen flow of 15 nl/hr. After activation the reactor was set at the initial temperature, pressurised and the feed was started.

Evaluation conditions:

Feed:	5 mixture of 10% tetralin in solvent D40, containing 25 ppm S as Benzothiophene
V catalyst:	5 ml
LHSV:	10 h ⁻¹
GHSV:	3000 h ⁻¹
Pressure:	30 bar H ₂
T inlet:	250 °C

Product analysis:

The aromatic concentration was measured with the UV-spectrophotometer at 274 nm.

Hydrogenation results and Discussion:

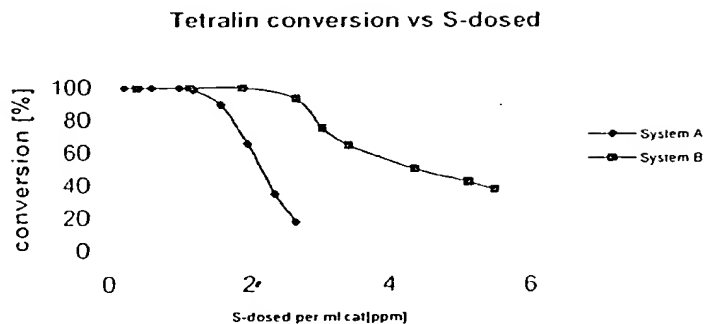


Figure 1: Tetralin conversion versus S-dosed. System A (cf. US patent 3,796,654); System B is 3-bed catalyst system.

In Figure 1, the tetralin conversion is depicted versus the amount sulphur dosed per volume catalyst. This figure shows that system B has a better activity performance than system A.

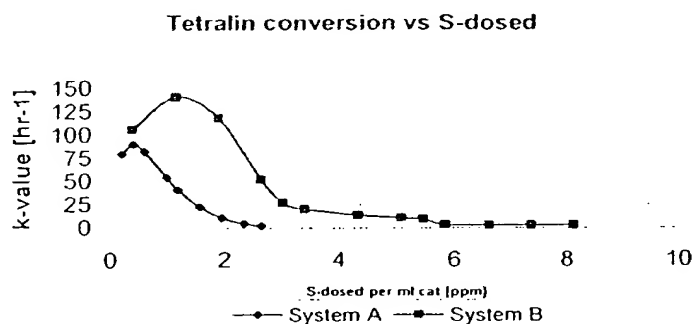


Figure 2: k-value versus S-dosed of the tetralin conversion. System A is UOP catalyst; System B is 3-bed catalyst system.

In Figure 2, the k-value is depicted versus the amount Sulphur dosed per volume catalyst. This figure shows that system B has a higher k-value than system A.

Further experiments were performed in an atmospheric gasphase hydrogenation apparatus. The feed consisted of tetralin in hydrogen. The hydrogen flow was 400 ml/min with approx. 0.035 vol.% tetralin. In these tests the fraction of tetralin converted at 230 °C was measured as function of the number of thiophene pulses dosed to the catalyst systems.

In each experiment the same amount of metals were loaded in the reactor. Loaded was 250 mg of catalyst A, the reactor load contained 0.94 mg Pt, 1.62 mg Sn and 12.5 mg Ni. The volume of this catalyst was approx. 0.75 ml.

Catalyst C consisted of three beds in series containing the following: 188 mg 0.5 wt.% Pt/Alumina (approx. 0.94 mg Pt), 40 mg 4.0 wt.% Sn catalyst (approx. 1.62 mg Sn) and 24 mg 56 wt.% Ni catalyst (approx. 12.5 mg Ni)

Catalyst D consisted of a physical mixture of the three catalysts of catalyst C with the same metal loading per reactor load

Catalyst E consisted of three beds in series containing the following:

187 mg 0.5 wt.% Pt/alumina (approx. 0.94 mg Pt), 39 mg 4.1 wt.% Mo/Fe (approx. 1.62 mg Mo/Fe) and 21 mg 56 wt.% Ni catalyst (approx. 12.5 mg Ni)

Tetralin conversion versus S-dosed

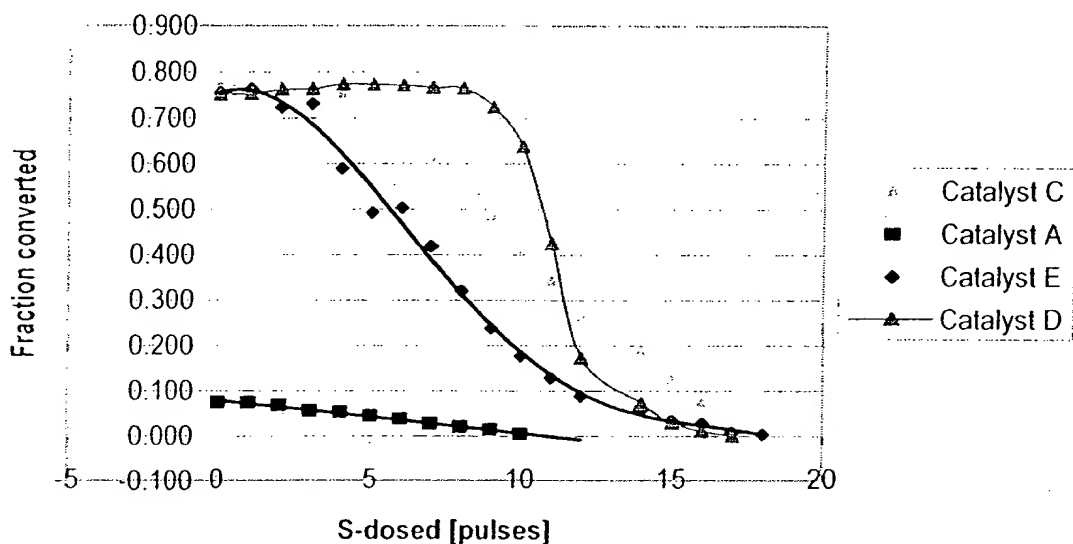


Figure 3 Tetralin conversion as function of the number of Sulphur dosages.

From the experiments it can be concluded that the performance of the claimed system is resulting in better performance than the catalytic composite of the process claimed in US 3,796,654.